

FORM PTO-1390  
(REV. 9-2001)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

OHS-314

U.S. APPLICATION NO. 10/019076 37 CFR 1.5

10/019076

INTERNATIONAL APPLICATION NO.  
PCT/JP00/02835INTERNATIONAL FILING DATE  
28 April 2000

PRIORITY DATE CLAIMED

TITLE OF INVENTION

METHOD FOR PRODUCING POLYPHENYLENE ETHER

APPLICANT(S) FOR DO/EO/US

Kiyoshi YOSHIDA, Naoto OHSAKI, Hiroya FUJII

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☐ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☐ is attached hereto (required only if not communicated by the International Bureau).
  - b. ☒ has been communicated by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a. ☒ is attached hereto.
  - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ have been communicated by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

## Items 11 to 20 below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☒ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☐ A FIRST preliminary amendment.
14. ☐ A SECOND or SUBSEQUENT preliminary amendment.
15. ☐ A substitute specification.
16. ☐ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☒ Other items or information: Copy of IB/308

U.S. APPLICATION NO. <b>10/019076</b> INTERNATIONAL APPLICATION NO. <b>PCT/JP00/02835</b>				ATTORNEY'S DOCKET NUMBER <b>OHS-314</b>	
21. <input checked="" type="checkbox"/> The following fees are submitted: <b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):</b> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... <b>\$1040.00</b>  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... <b>\$890.00</b>  International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... <b>\$740.00</b>  International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$710.00</b> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... <b>\$100.00</b> <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>CALCULATIONS PTO USE ONLY</b>          <div style="border: 1px solid black; padding: 2px;">\$ 890.00</div>	
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CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	8 - 20 =	0	x \$18.00	\$ 0	
Independent claims	1 - 3 =	0	x \$84.00	\$ 0	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$280.00	\$ 0	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$ 890.00	
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U.S. APPLICATION NO. (15 USC § 371) 10/019076

INTERNATIONAL APPLICATION NO.  
PCT/JP00/02835ATTORNEY'S DOCKET NUMBER  
OHSB-31421. ☒ The following fees are submitted:**BASIC NATIONAL FEE** (37 CFR 1.492 (a) (1) - (5)):Neither international preliminary examination fee (37 CFR 1.482)  
nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO  
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\$740.00

International preliminary examination fee (37 CFR 1.482) paid to USPTO  
but all claims did not satisfy provisions of PCT Article 33(1)-(4) .....

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CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$
Total claims	8 - 20 =	0	x \$18.00	\$ 0
Independent claims	1 - 3 =	0	x \$84.00	\$ 0
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Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$ 0
<b>TOTAL NATIONAL FEE =</b>				\$ 890.00
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Total claims

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Independent claims

1 - 3 =

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MULTIPLE DEPENDENT CLAIM(S) (if applicable)

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**TOTAL OF ABOVE CALCULATIONS =**

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19,636  
REGISTRATION NUMBER

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2001年11月8日 (08.11.2001)

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- (51) 国際特許分類: C08G 65/46, 65/44, C08J 3/14 Naoto) [JP/JP]. 藤井弘也 (FUJII, Hiroya) [JP/JP]; 〒510-0886 三重県四日市市日永東2丁目4番16号 三菱瓦斯化学株式会社 四日市工場内 Mie (JP).
- (21) 国際出願番号: PCT/JP00/02835
- (22) 国際出願日: 2000年4月28日 (28.04.2000) (74) 代理人: 弁理士 大島正孝 (OHSHIMA, Masataka); 〒160-0004 東京都新宿区四谷四丁目3番地 福屋ビル 大島特許事務所 Tokyo (JP).
- (25) 国際出願の言語: 日本語
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- (71) 出願人 (米国を除く全ての指定国について): 三菱瓦斯化学株式会社 (MITSUBISHI GAS CHEMICAL COMPANY, INC.) [JP/JP]; 〒100-0005 東京都千代田区丸の内2丁目5番2号 Tokyo (JP). (84) 指定国 (広域): ヨーロッパ特許 (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).
- (72) 発明者; および
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(54) Title: PROCESS FOR PRODUCING POLYPHENYLENE ETHER

(54) 発明の名称: ポリフェニレンエーテルの製造法

(57) Abstract: A phenol is oxidatively polymerized in an aromatic compound solvent in the presence of a copper compound and an amine to produce a polyphenylene ether resin, during which the aromatic compound solvent, the amine forming an azeotropic mixture together with water, and the methanol used for precipitating polyphenylene ether particles and for washing the particles are efficiently separated and recovered.

(57) 要約:

フェノール類を芳香族化合物溶媒中、銅化合物およびアミン類の存在下で酸化重合させてポリフェニレンエーテル樹脂を製造するに際して、芳香族化合物溶媒、水と共沸混合物を作るアミン類、並びに、ポリフェニレンエーテル粒子の析出および該粒子の洗浄に用いるメタノールを効率よく分離回収する。

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WO 01/83587 A1

## DESCRIPTION

## METHOD FOR PRODUCING POLYPHENYLENE ETHER

5 Technical Field

The present invention relates to a method for producing polyphenylene ether. More specifically, the present invention relates to a method for producing the polyether that includes recycle use of a solvent.

10 Prior Art

As a method for synthesizing the polyphenylene ether, there is a method of subjecting phenol to oxidative polymerization in the presence of a copper compound and amines such as di-n-buthylamine and the like, in an aromatic compound solvent. A non-solvent for the polyphenylene ether such as methanol, water or the like is added in the above-obtained polyphenylene ether solution to precipitate the polyphenylene ether particles. Then, dry particles of the polyphenylene ether can be prepared through steps such as solid-liquid separation, washing with methanol or the like and dryness.

It surely needs to achieve reduction of the manufacture cost that components used in the above method of preparing the polyphenylene ether, such as the aromatic compound solvent, the amines, methanol or water, are each separated and recovered for every component to reuse. However, when di-n-buthylamine is used especially as the amines, it has not been established a method of separating and recovering an aromatic compound solvent, di-n-buthylamine and methanol efficiently and economically.

The present invention has been made to solve the above problems, and the object of the present invention is to provide an industrially advantageous method for producing the polyphenylene ether by separating and recovering efficiently the solvent used in polyphenylene ether production process,

such as aromatic compound solvent, amines and methanol.

Other purposes and advantages of the present invention will become clear from the following explanation.

Disclosure of invention

5           The present inventors have made extensive studies to solve the above problems, and as a result, have found that it was able to recover methanol having the low amines content and water by adding water to a mixed solution of the aromatic compound solvent, amines and methanol, separating a phase  
10 which consists of the aromatic compound solvent and a large portion of the amines from the mixed solution by liquid-liquid separation, supplying a phase which consists mainly of methanol containing a part of the amines and water to a distillation column, and distilling the phase while carrying  
15 out side-cut of the amines. Thus, the present invention has been accomplished.

That is, the present invention is a method for preparing polyphenylene ether comprising the following steps,

(a) a step for synthesizing polyphenylene ether by  
20 subjecting phenol to oxidative polymerization in the presence of a copper compound and amines in an aromatic compound solvent,

(b) a step for precipitating the polyphenylene ether particles by adding methanol to the solution of polyphenylene ether in the aromatic compound solvent obtained by the step  
25 (a),

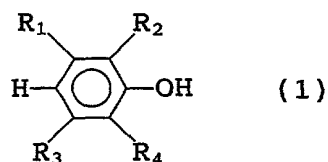
(c) a step for solid-liquid separating a slurry of polyphenylene ether obtained by the step (b), and washing the separated polyphenylene ether particles with methanol to obtain the polyphenylene ether,

30           (d) a step for adding water to a filtrate obtained by the step (c) to mix them, and then, liquid-liquid separating the mixture into phase consisting mainly of the aromatic compound and a phase consisting mainly of methanol and water, and

(e) a step for supplying the phase consisting mainly of methanol and water, obtained by the step (d), to the middle portion of the distillation column to perform distillation, and separating out a distillate liquid consisting mainly of methanol, a bottom liquid consisting mainly of water and a side-cut liquid comprising the amines by distilling, recycling the distillate liquid consisting mainly of methanol as methanol which is used at the step (b) and (c), and recycling the side-cut liquid as part of the filtrate which is used at the step (d).

Embodiment for practicing the invention

The polyphenylene ether in the present invention is derived from a compound represented by the following formula (1) by oxidative polymerization.



(wherein  $\text{R}_1$ ,  $\text{R}_2$ ,  $\text{R}_3$  and  $\text{R}_4$  represent independently a substituent selected from the group consisting of a hydrogen atom, an alkyl group, a substituted alkyl group, a halogen atom, an aryl group and a substituted aryl group.)

Examples of polyphenylene ether according to the present invention are a homopolymer such as

poly(2,6-dimethyl-1,4-phenylene)ether,

poly(2-methyl-6-ethyl-1,4-phenylene)ether,

poly(2,6-diethyl-1,4-phenylene)ether,

poly(2-ethyl-6-n-propyl-1,4-phenylene)ether,

poly(2-methyl-6-n-propyl-1,4-phenylene)ether,

poly(2-ethyl-6-isopropyl-1,4-phenylene)ether,

poly(2-methyl-6-chloroethyl-1,4-phenylene)ether,

poly(2-methyl-6-hydroxyethyl-1,4-phenylene)ether;

polyphenylene ether copolymer such as 2,6-dimethylphenol copolymerized with 2,3,6-trimethylphenol and/or o-cresol.

The step (a) of the present invention is a step for obtaining the polyphenylene ether by subjecting phenol to oxidative polymerization in the presence of the copper compound and the amines in the aromatic hydrocarbon solvent.

5 The aromatic hydrocarbon solvent used in the reaction of polymerization is preferably benzene, toluene and xylene, and especially toluene is preferred. The concentration of polyphenylene ether in a solution of polymerization reaction is preferably 5 to 70 wt%, more preferably 10 to 50 wt%. Further,  
10 the solution may contain substances for removing catalysts, by-products or the like, other than polyphenylene ether and catalysts for polymerization (the copper compound and the amines).

The step (b) is a step for precipitating the  
15 polyphenylene ether particles by adding methanol which is a non-solvent for polyphenylene ether, to the solution of polyphenylene ether in the aromatic compound solvent, obtained by the step (a).

The amount of methanol to be added is preferable 1 to  
20 2 times by weight of the aromatic compound in polyphenylene ether solution. It is desirable further to let water co-exist with a mixture of the aromatic compound solvent and methanol within such a range that the mixture can maintain a single liquid phase. Moreover, when the solution of polymerization  
25 reaction is supplied, it is preferred that the polyphenylene ether solution and methanol are continuously supplied into one or more stages of a stirring vessel having a reciprocating stirrer. Further, to decrease the amount of methanol used, it is desirable to supply the polyphenylene ether solution  
30 into the stirring vessel while maintaining temperature to the extent that polyphenylene ether does not precipitate after heating and condensing the polyphenylene ether solution.

The step (c) is a step for solid-liquid separating the slurry of polyphenylene ether obtained by the step (b), and



then, washing the separated polyphenylene ethers particles with methanol.

As a method of solid-liquid separating the slurry of the polyphenylene ether particles precipitated, for example, there are a method of using a centrifugal force and a vacuum filtration method under reduced pressure. Moreover, not only these but any methods can be used. The solid-liquid separated wet particles of polyphenylene ether were washed with methanol, and dried by heating or the like, to obtain dry particles. Since it is hard to remove the aromatic compound solvent and the amines by drying, if these are not removed from the wet particles by methanol-washing, these will remain in the dry particles and will become cause a stench or the like.

The step (d) is a step for adding water to the filtrate obtained by the step (c), i.e. the filtrate containing the aromatic compound, the amines and methanol, and liquid-liquid separating into a phase consisting mainly of the aromatic compound and a phase consisting mainly of methanol and water.

It is desirable to set up the amount of water to be added so that the weight ratio of water/methanol in the liquid-liquid separated raw solution may be 0.5 to 1.5. Further, as for a method of liquid-liquid separation, there are a stand separation, a centrifugation and a tower system separation or the like. Moreover, not only these but any methods can be used. After most of the aromatic compound and the amines are recovered as the phase consisting mainly of the aromatic compound, if desired, it is fractionated to recycle in a polymerization reaction process.

The step (e) is a step for supplying the phase consisting mainly of methanol and water, obtained by the step (d), i.e. the phase consisting mainly of methanol containing amines, and water to the middle portion of the distillation column, and separating out the distillate liquid consisting mainly of methanol, the bottom liquid mainly consisting of water

and the side-cut liquid containing the amines. The phase consisting mainly of methanol and water, to be distilled preferably contains 40 to 60 wt% of methanol, 40 to 60 wt% of water and 0.1 to 0.3 wt% of the amines.

5        Though both a tray type column and a packed column can be used as the distillation column, the tray type column is desirable because of ease of a side-cut. Moreover, there is no particular restriction in distillation pressure.

10        The distillate solution consisting mainly of methanol, obtained by distillation is recycled to be used as the non-solvent for precipitating polyphenylene ether in the step (b) or as the liquid for washing the wet particles of polyphenylene ether in the step (c). If the amines in methanol used as the liquid for washing in the step (c) are contained  
15        in a high percentage, it will become inadequate to remove the amines in the polyphenylene ether particles. Therefore, it is preferable that the concentration of the amines in the distillate methanol is less than 0.1 wt%.

20        The bottom liquid consisting mainly of water is recycled as water used in the step (d). To remove the accumulated impurities, it is preferable to discharge at least part of water out of the system. In this case, it is the simplest way to discharge water as drainage, however, when water contains a high percentage of the amines which becomes a source  
25        of COD in drainage, drainage disposal will be complicated. Moreover, since the cost increases due to the supplement of the discharged water, it is preferable that the concentration of the amines in the bottom liquid is less than 0.01 wt%.

30        To maintain the concentration of the amines in the distillate solution and bottom solution less than a certain concentration in the step (e), the solution containing the amines is extracted from the middle portion of the distillation column as the side-cut liquid. It is desirable to set the position for extracting the side-cut liquid so that the

concentration of the amines in the side-cut liquid becomes as high as possible. Further, it is desirable to set the extracted amount so that it is 0.5 to 5wt.% of the phase consisting mainly of methanol and water, to be distilled.

5 The side-cut liquid is returned to the step (d). Therefore, the most of the amines distributed to the phase consisting mainly of methanol and water in the step (d) circulates between the step (d) and (e), and as a result, the amines which are discharged as the bottom liquid can be held down in a very  
10 little.

For the purpose of effective distilling and the like, an anti-foaming agent or sodium hydroxide can be added into the phase consisting mainly of methanol and water, to be distilled in the step (e). In this case, to avoid returning  
15 the additive to the step (d), it is desirable to carry out side-cut from between the tray supplying a material solution and the top of the distillation column.

Examples of the amines of the present invention include the amines making an azeotrope with water such as  
20 di-n-buthylamine, n-buthyldimethylamine and the like.

#### Examples

The following examples illustrate the present invention more specifically, but are in no way to be taken as limiting  
25 the invention.

#### Example 1

Two kilograms of cupric bromide was dissolved in 35kg of di-n-buthylamine and 800kg of toluene. A solution in which 200kg of 2,6-dimethylphenol was dissolved in 500kg of toluene  
30 was added to this catalytic solution. This mixed solution was subjected to polymerization at 40°C for 3 hours while supplying oxygen into a reactor. After the reaction stopped, the solution was contacted with water to remove the catalyst from the reaction solution, and the polymerization reaction

solution of polyphenylene ether was obtained. This polyphenylene ether solution was mixed while stirring with methanol of 1.5 times by weight of toluene in the solution, to precipitate the polyphenylene ether particles. Then, the solution was separated by a solid-liquid separator, and a wet solid was washed with methanol of 5 times by weight of polyphenylene ether in the wet solid. Water was added to the solid-liquid separated filtrate so that the weight ratio of water/methanol after addition might be 1.0, and the solution was subjected to stand separation to obtain a phase of toluene and a phase consisting mainly of methanol and water. When distillation was performed, while carrying out side-cut, by using this phase consisting mainly of methanol and water as a material under conditions shown in Table 1, the distillate and the bottom liquid having the low concentration of di-n-buthylamine was obtained. The result was shown in Table 1.

Table 1

	Stages	Flow Rate (g/h)	Composition(wt%)			
			ME	TL	DBA	water
Supplied Liquid	20	661	50.1	1.7	0.2	48.0
Distilled Liquid	1	338	95.3	3.4	0.02	1.3
Bottom Liquid	40	305	0.02	<0.01	<0.01	99.98
Side-cut Liquid	15	13	43.2	0.1	9.6	47.1

ME: methanol

TL: toluene

DBA: di-n-buthylamine

#### Comparative Example 1

An experiment was carried out under the same conditions as Example 1 except that distillation was performed, without carrying out side-cut, by using the phase consisting mainly of methanol and water as a material under conditions shown in Table 2. The concentration of di-n-buthylamine in the

distillate liquid and bottom liquid obtained by distillation, increased remarkably as compared with Example 1. In addition, the temperature of each stage of the distillation column was also unstable. The result was shown in Table 2.

5

Table 2

	Stages	Flow Rate (g/h)	Composition (wt%)			
			ME	TL	DBA	water
Supplied Liquid	20	655	50.1	1.7	0.2	48.0
Distillate Liquid	1	335	95.4	3.1	0.18	1.3
Bottom Liquid	40	320	0.04	<0.01	0.22	99.74

ME: methanol

TL: toluene

10 DBA: di-n-buthylamine

According to the present invention, the aromatic compound solvent used for the production of polyphenylene ether, the amines making the azeotrope with water, and methanol  
 15 can be separated and collected efficiently, and polyphenylene ether having low concentration of residual solvent can be produced at lower cost.

## CLAIMS

1. A method for producing polyphenylene ether, which comprises
- 5 (a) a step for synthesizing polyphenylene ether by subjecting phenol to oxidative polymerization in the presence of a copper compound and amines in an aromatic compound solvent,
- (b) a step for precipitating polyphenylene ether particles by adding methanol into the solution of polyphenylene ether
- 10 in the aromatic compound solvent obtained by the step (a),
- (c) a step for solid-liquid separating a slurry of polyphenylene ether obtained by the step (b), and then washing the separated polyphenylene ether particles with methanol to obtain the polyphenylene ether,
- 15 (d) a step for adding water to a filtrate obtained by the step (c) to mix them, and then liquid-liquid separating the mixture into a phase consisting mainly of the aromatic compound and a phase consisting mainly of methanol and water, and
- (e) a step for supplying the phase consisting mainly of methanol and water, obtained at the step (d) to the middle portion
- 20 of a distilled column to perform distillation, and separating out a distillate liquid consisting mainly of methanol, a bottom liquid consisting mainly of water and a side-cut liquid containing the amines by distillation, recycling the
- 25 distillate liquid consisting mainly of methanol as methanol of the steps (b) and (c), and recycling the side-cut liquid as part of the filtrate of the step (d).
2. The method of claim 1, wherein the amines are amines
- 30 which make an azeotrope with water.
3. The method of claim 1, wherein the amount of methanol added at the step (b) is 1 to 2 times by weight of the aromatic compound in the solution.

4. The method of claim 1, wherein water is added so that the weight ratio of water/methanol in the liquid-liquid separated raw material solution in the step (d) is 0.5 to 1.5.

5. The method of claim 1, wherein the phase consisting mainly of methanol and water, to be distilled in the step (e) contains 40 to 60 wt% of methanol, 40 to 60 wt% of water and 0.1 to 0.3 wt% of the amines.

6. The method of claim 1, wherein the amount of side-cut liquid is 0.5 to 5 wt% based on the phase consisting mainly of methanol and water, to be distilled.

7. The method of claim 1, the concentration of the amines in the distillate solution in the step (e) is less than 0.1 wt% and the concentration of the amines in the bottom liquid is less than 0.01 wt%.

8. The method of claim 1, wherein an anti-foaming agent and/or sodium hydroxide are/is added to the phase consisting mainly of methanol and water, to be distilled in the step (e), and carrying out side-cut between the stage supplying a raw material and the top of the distillation column.

Dkt. No. OHS-314**DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION**

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (*if only one name is listed below*) or an original, first and joint inventor (*if plural names are listed below*) of the subject matter which is claimed and for which a patent is sought on the invention entitledMETHOD FOR PRODUCING POLYPHENYLENE ETHER\_\_\_\_\_, the specification of which:  
(check one) ☐ is attached hereto ☒ was filed on April 28, 2000 as International Application Serial No. PCT/JP00/02835 and was amended on \_\_\_\_\_ (*if applicable*).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(a) of any PCT international application which designated at least one country other than the United States, listed below and have also identified below any foreign application for patent or inventor's certificate, or PCT international application having a filing date before that of the application on which priority is claimed:

## Prior Foreign Application(s)

## Priority Claimed

(NUMBER) _____	(COUNTRY) _____	(FILED D/M/Y) _____	<input type="checkbox"/> <input type="checkbox"/> YES NO
(NUMBER) _____	(COUNTRY) _____	(FILED D/M/Y) _____	<input type="checkbox"/> <input type="checkbox"/> YES NO
(NUMBER) _____	(COUNTRY) _____	(FILED D/M/Y) _____	<input type="checkbox"/> <input type="checkbox"/> YES NO
(NUMBER) _____	(COUNTRY) _____	(FILED D/M/Y) _____	<input type="checkbox"/> <input type="checkbox"/> YES NO
(NUMBER) _____	(COUNTRY) _____	(FILED D/M/Y) _____	<input type="checkbox"/> <input type="checkbox"/> YES NO
(NUMBER) _____	(COUNTRY) _____	(FILED D/M/Y) _____	<input type="checkbox"/> <input type="checkbox"/> YES NO
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I hereby claim the benefit under 35 U.S.C. §120 of any United States application(s), or §365(c) of any PCT international application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of 35 U.S.C. §112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(APPLICATION SERIAL NO.) _____	(FILING DATE) _____	(STATUS) _____
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**POWER OF ATTORNEY:** As a named inventor, I hereby appoint the following attorneys and/or agents to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine and imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or document or any patent issuing thereon.

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